# A cluster model of spin glasses: Towards reconciling theory and experiment

K. Levin, C. M. Soukoulis,[a)] and G. S. Grest

*James Franck Institute, University of Chicago, Chicago, Illinois 60637*

We have proposed a cluster mean field theory of the spin glasses in which dynamical clusters of spins are the fundamental entity. We review experimental evidence for spin clusters and show that within our model it is possible to understand (i) why the specific heat and resistivity have a broad maximum at temperatures above $T_{SG}$ at which the magnetic susceptibility cusp appears and (ii) why the total neutron cross section $d\sigma/d\Omega_k$ has a k-dependent maximum at temperatures below $T_{SG}$, where k is the momentum transfer to the neutron. These results suggest that there may, in fact, be a sharp transition to the spin glass phase, which turns out to be more apparent in susceptibility than in specific heat, resistivity, and neutron measurements.

PACS numbers: 64.60.Cn, 75.10.Hk, 75.30.Cr, 75.40. – s

The purpose of this paper is to offer a theoretical explanation for a number of experimental results in the spin glass alloys, within the context of a cluster mean field theory [1-3]. For the most part these experiments have been interpreted by others as suggesting that there is no sharp phase transition in the spin glasses. We argue here that this is not necessarily the case and show that they can be reconciled with the notion that the alloy undergoes a sudden freezing in the spins at a single temperature $T_{SG}$. We will discuss specific heat [4], neutron scattering [5,6], resistivity [7,8] and finite field (reversible) susceptibility and magnetization measurements [9-14]. See [15, 16] for a more complete review of the experimental results. Because a theory for the first two of these is discussed in detail elsewhere [1-3], attention here will be focused on the behavior of the resistivity and finite field measurements.

Our theoretical framework is based on a natural merger of two central ideas in the spin glasses: the Edwards-Anderson (EA) mean field theory [17] and the notion that clusters rather than individual spins are the fundamental entity. The former has been discussed in detail elsewhere [18-20] and needs no review here.

Different cluster models of the spin glasses have been formulated by a number of researchers. In 1971, Beck [10] first pointed out that short range ferromagnetic order is present in Cu-Mn and similar alloys. This observation was based on the "superparamagnetism" evident from measurements of the field and temperature dependence of the magnetization. In addition to ferromagnetic clusters, he argued that one had to assume a matrix of much smaller moments in which the spin directions are frozen below some temperature $T_{SG}$. Due to the interaction of the giant moments and the frozen matrix, the spins of the cluster gradually become frozen as the temperature is decreased. Tholence and Tournier [12] based their arguments for the existence of spin clusters on analogies with the Néel theory of rock magnetism. The alloy is spontaneously divided into independent regions (monodomains). The exchange interactions within each region are given by the RKKY form. At T = 0 the resulting moment (due to imperfect compensation of the spins) is frozen in the direction of its random anisotropy axis. This model was used to correlate measurements of the saturated magnetization $\sigma_s$, the zero temperature saturated remanent magnetization $\sigma_{rs}(0)$ and the thermal variation of $\sigma_{rs}$.

Binder [21] suggested that ferromagnetic clusters of spins must be invoked (among other reasons) in order to understand the magnitude of the remanent magnetization and the field sensitivity of the AC (reversible) susceptibility measurements. He noted that a simple EA theory yielded disagreement with experimental measurements of these quantities by at least an order of magnitude. This could be corrected by rescaling the spin size S to a larger value (presumably derived from large rigid clusters). Murani [6] based his argument for the existence of clusters on his neutron scattering experiments. He observed that the $\frac{d\sigma}{d\Omega_k}$ had a maximum at a temperature which depended on the momentum k and therefore suggested that this reflected the freezing temperature of a cluster of characteristic size 1/k. Since this temperature was k-dependent he argued that the clusters must freeze along their random anisotropy axes at different temperatures depending on their characteristic size. Additional cluster models similar to those summarized here have been discussed by Mydosh [15], Guy [13], Coles et al. [22], and Kouvel [23].

Independently of Binder [21], two of the present authors [1] suggested a self-consistent cluster model based on the EA theory. Unlike that discussed by Binder, the model treated exactly the internal degrees of freedom of the cluster, thus enabling us to discuss the temperature dependence of various quantities. Briefly the model can be summarized as follows. We begin with a phenomenological model Hamiltonian,

$$\mathcal{H} = -\sum_{\nu<\lambda} J_{\nu\lambda} \vec{S}_\nu \cdot \vec{S}_\lambda + \sum_\nu \sum_{i<j} J_{ij}^{o} \vec{S}_{i\nu} \cdot \vec{S}_{j\nu} - g\mu_B \sum_{\nu i} \vec{H} \cdot \vec{S}_{i\nu} \qquad (1)$$

where $\vec{S}_{i\nu}$ denotes the spin at the site i in the $\nu^{th}$ cluster and $\vec{S}_\nu = \sum_i \vec{S}_{i\nu}$. $J_{ij}^{o}$ is the intra-cluster exchange which is treated exactly. The inter-cluster exchange interactions $J_{\mu\nu}$, treated in mean field theory, are distributed according to the Gaussian formula

$$P(J_{\nu\lambda}) = 1/[(2\pi)^{1/2} J] \exp [-(J_{\nu\lambda} - J'_{\nu\lambda})^2/2J^2], \qquad (2)$$

where J' is the net ferromagnetic intercluster exchange inter-

 0021-8979/79/031695-05$01.10  

action. The free energy is evaluated using the replica method in terms of the variational parameters $Q = [\langle \vec{S}_\nu\rangle \cdot \langle \vec{S}_\nu\rangle]_c$, $M = [\langle \vec{S}_\nu \cdot \vec{S}_\nu\rangle]_c$ and $\vec{m} = [\langle \vec{S}_\nu\rangle]_c$, where $[\,]_c$ denotes a configuration average. This yields for Heisenberg spins

$$F(q,M,m) = -N^{cl}kT\,[\frac{\bar{J}^2}{12(kT)^2}(Q^2-M^2) - \sum_{\lambda\neq\nu}\frac{J'_{\nu\lambda}\,\vec{m}\cdot\vec{m}}{2(kT)}$$

$$+ (2\pi)^{-3/2}\int d^3r\, e^{-r^2/2}\,\ln \mathrm{Tr}\exp(-\beta H^{eff})] \quad (3)$$

and

$$-\mathcal{H}^{eff} = \sum J^{o}_{ij}\vec{S}_{i\nu}\cdot\vec{S}_{j\nu} + \bar{J}(Q/3)^{1/2}\vec{r}\cdot\vec{S}_\nu + \frac{\bar{J}^2}{6kT}(M-Q)\vec{S}_\nu\cdot\vec{S}_\nu$$

$$+\vec{S}_\nu\cdot\sum_{\lambda\neq\nu}J'_{\nu\lambda}\vec{m} + g\mu_B\vec{H}\cdot\vec{S}_\nu \quad (4)$$

where the sums involving J' include only near neighbor pairs of spins. Here $\bar{J} = zJ$, where z is the number of nearest neighbors and $N^{cl}$ is the number of clusters.

In mean field theory, each cluster can be assumed to contain the same number of spins, which is equal to the average value in the alloy. The magnetic susceptibility and specific heat are readily evaluated. The former is most sensitive to the random spin glass ordering between the clusters rather than to the short range order within the cluster. Therefore it does not differ significantly from what is found in the single spin EA case, and is in reasonable agreement with experiment. The latter primarily reflects the intracluster short range order. Although it has a weak contribution from the spin glass freezing, most of this is washed out by the Schottky anomaly arising from the spin cluster. This leads to a broad maximum in $C_m$ at a temperature somewhat higher than the freezing temperature. This cluster model thus "corrects" a basic defect of the EA picture by building in short range order at $T_{SG}$ and thereby yielding a specific heat which is qualitatively in agreement with experiment.

A second success of the model was a consistent interpretation [2] of Murani's neutron scattering experiments. We showed elsewhere that these experiments are, in fact, consistent with the notion that the spin glasses undergo a sharp phase transition rather than a continuous one as has been suggested [6]. We presented two arguments to support our claim. The first was a simple data analysis. In this we demonstrated that the total experimental neutron cross section, which consists of the two terms

$$\frac{d\sigma}{d\Omega_k} \propto \sum_k e^{i\vec{k}\cdot\vec{R}_{ij}}\{[\langle\vec{S}_i\cdot\vec{S}_j\rangle]_c - [\langle\vec{S}_i\rangle\cdot\langle\vec{S}_j\rangle]_c\}$$

$$+\sum_k e^{i\vec{k}\cdot\vec{R}_{ij}}[\langle\vec{S}_i\rangle\cdot\langle\vec{S}_j\rangle]_c \quad (5a)$$

$$\propto T\chi(k,T) + I_B(k,T) \quad (5b)$$

can be decomposed into these separate contributions. The second term, called the "Bragg" term by analogy with ferromagnetism is basically a Fourier transform of the order parameter. It thus vanishes for $T \geq T_{SG}$. Its zero temperature contribution is given by the total cross section at $T = 0$, since $T\chi(k,T)$ vanishes there. We have a reasonably good idea of the shape of the Bragg curve from the expected T dependence of the order parameter. The corresponding curves are shown in the insets of Figs. 1a, 1b for two different AuFe alloys [6] containing 13 and 15% Fe. The net result of subtracting the Bragg term (dashed) from the total cross section yields $T\chi(k,T)$ which is plotted in the main part of both figures for a range of k values. As can be seen these curves all have their maxima at a single temperature $T_{SG}$ (taken to be 40 K and 43 K in the 13 and 15% sample, respectively). The data analysis thus suggests the existence of a sharp phase transition rather than a continuous one. What we have shown is that this transition is reflected in $\chi(k,T)$,

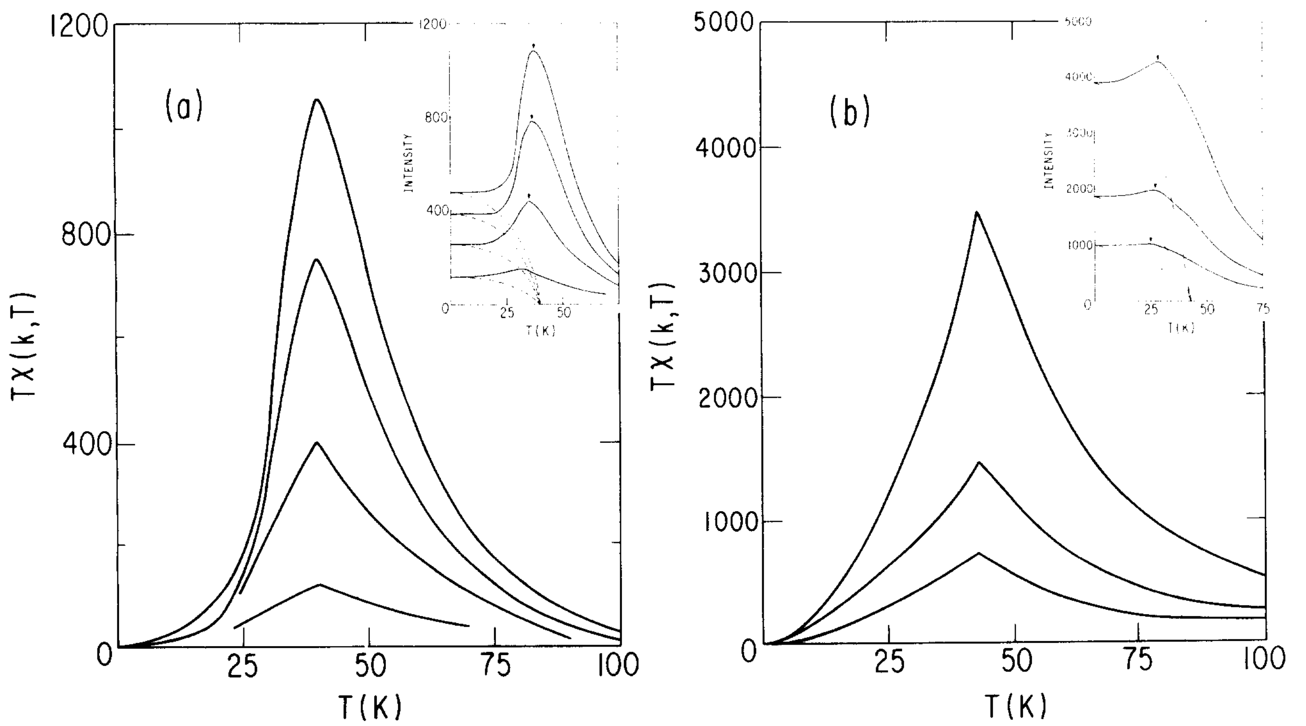



Fig. 1 The wave vector dependent susceptibility $T\chi(k,T)$ in a AuFe alloy for (a) 13% and (b) 15% Fe deduced by subtracting an estimate of the "Bragg" term (dashed) from the measured (Ref. 6) total cross section I(k, T) (plotted in the inset). The value of q from top to bottom are (a) 6.0, 6.9, 8.6 and 13.8 x $10^{-3}$ Å$^{-1}$ for 13% Fe and (b) 5.2, 6.9 and 8.6 for 15% Fe.

but not in the total neutron cross section. The behavior is in contrast to what is observed in a pure ferromagnet for small k, where the two are essentially equivalent.

We have performed a simple RPA calculation of $\frac{d\sigma}{d\Omega_k}$, $T\chi(k, T)$ and $I_B(k, T)$ using the cluster mean field theory. The result of this calculation [2] is that $\frac{d\sigma}{d\Omega_k}$ has a k-dependent peak for $T < T_{SG}$, whenever (i) the inter-cluster ferromagnetic exchange constant $J' \neq 0$, corresponding to the shift of the Gaussian and (ii) $\frac{dM}{dT} \neq 0$. An illustrative example of these effects is in Ref. 2.

There are two different indications that clusters of spins are important in these neutron experiments. The first comes from the observation that the Bragg curves, derived from experiment at T = 0 are strongly k dependent. Thus the order parameter $[\langle S_i \rangle \cdot \langle S_j \rangle]_c$ is not simply proportional to $\delta_{ij}$ as in the EA theory. Secondly, in a Heisenberg EA model, M will be a constant in temperature. Hence in this theory which ignores clusters dM/dT = 0 and the maximum in the cross section occurs at $T = T_{SG}$ in contradiction to experiment [6].

The characteristic behavior of the resistivity $\rho$ in a spin glass is shown in Fig. 2a. The data is taken from Ref. 7. As can be seen there is a broad maximum in $\rho$ at a temperature above $T_{SG}$ and generally somewhat larger than that of the specific heat maximum. The qualitative shape of these curves has been explained previously by Beal-Monod [24] and Larsen [25]. The former author considered the perturbative contributions to the resistivity (up to third order in the s-d exchange constant $J^{sd}$) from pairs of spins; the spin glass transition was not explicitly considered. The latter used a noise model, which presupposes the existence of internal magnetic fields above $T_{SG}$, and summed diagrams in $J^{sd}$ within the parquet approximation. We point out here that only by introducing spin clusters (with internal degrees of freedom), can the data be reconciled with a theory of a mean field-like phase transition at $T_{SG}$. There are basically three temperature regions to distinguish: $0 \leq T \leq T_{SG}$, $T_{SG} \leq T \leq T_D$, and $T_D \leq T$, where $T_D$ is the degeneracy temperature of the spin cluster. This temperature is of order $\sim$ 2-3 $J^o$ where $J^o$ is the characteristic intra-cluster exchange temperature. We consider here only the second $\rho_2$ and third $\rho_3$ order contributions in $J^{sd}$. Our calculation of $\rho_2$ follows that of Levin-Mills [26]. We consider ferromagnetic clusters only. For $T \lesssim T_{SG}$, the temperature dependence of the resistivity is dominated by $\rho_2$, since the giant spin Kondo term $\rho_3$ is effectively quenched in an internal magnetic field. Because the cluster moment is relatively constant over this temperature region, to a good approximation $\rho_2$ and $\rho_3$ can be calculated by considering the magneto-resistivity of a giant spin. The former increases monotonically with T. In the intermediate temperature region $\rho_2$ continues to rise. This follows because the increasingly available inelastic processes dominate the effect of the decrease in the cluster moment. To some extent this can be understood as arising from interference effects due to the spatial extent of the cluster [26]. Above $T_D$, $\rho_2$ is a constant. For $T_D \lesssim T$, $\rho_3$ is simply the single spin Kondo term which decreases with temperature logarithmically. The overall effect of $\rho_2 + \rho_3$ is, then, a negative temperature coefficient of the resistivity, as observed in this temperature interval. For $T_{SG} \lesssim T \lesssim T_D$ the third order term $\rho_3$ increases with temperature. This is shown explicitly in Ref. 24. It can be understood as arising from the fact that the low T giant spin Kondo term is strongly suppressed by interference effects [26] (due to the spatial extent of the cluster). Hence between $T_{SG}$ and $T_D$, $\rho_3$ must smoothly extrapolate from a small $\ell n T$ term to a considerably larger one and increases monotonically. The results of a semi-quantitative calculation of $\rho$ are shown in Fig. 2b for a 10 spin cluster [27]. In the intermediate temperature region we have performed a smooth extrapolation, following the 2 spin results of Beal-Monod [24]. This places the maximum in $\rho$ at or below $T_D$. Note that this explains why this temperature is somewhat higher than the maximum in the specific heat which must necessarily occur well below $T_D$. If clusters are ignored, as in the simple EA picture, then it follows that $\rho$ must decrease for $T \gtrsim T_{SG}$ (in contradiction to experiment), since $\rho_2$ is constant there and $\rho_3$ decreases monotonically. Hence the behavior of the resistivity provides further evidence for spin clusters.

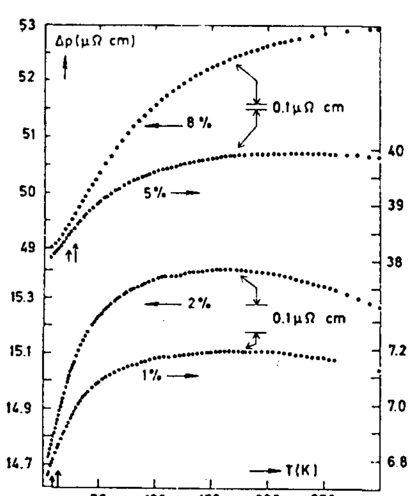
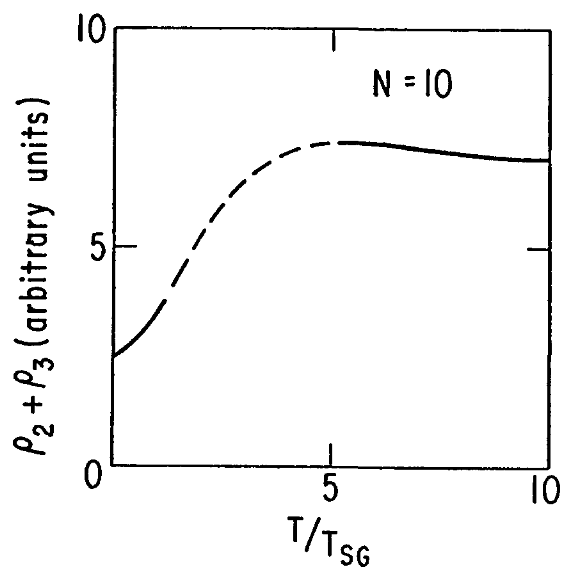



Fig. 2 (a) Over-all temperature variation of $\Delta\rho$ ($\mu\Omega$cm) for AuFe alloys with concentrations of 1-, 2-, 5-, and 8-at. % Fe. Note the change in scale between 1- and 2-at. % alloys, and the 5- and 8-at. % alloys. Data from Ref. 7.




(b) Resistivity $\rho = \rho_2 + \rho_3$ versus temperature for 10 spins in a cluster [27]. We have calculated $\rho$ for $T < T_{SG}$ and $T > T_D$ and the results are shown as a solid line. The intermediate region where we extrapolate between the two known limits is dashed.

As a final application of our cluster mean field theory we compute the field dependence of $\chi$, $C_m$ and the magnetization m for a range of temperatures. As was argued by Binder [21], the strong field sensitivity of $\chi$ and m suggests that spin clusters are important. It should be pointed out that there are both reversible and irreversible contributions to the magnetization and susceptibility below $T_{SG}$. Tholence and Tournier [12] showed that AC and DC measurements of $\chi$ were not equivalent. The former corresponds to the reversible contribution and exhibits a sharp cusp at $T_{SG}$ which disappears in a DC measurement. In magnetization studies [12, 13, 16], it was found that the history of the sample is very important and that results differ depending upon whether it was field cooled or not. It appears that all reversible magnetization and susceptibility measurements can be qualitatively explained by the EA mean field theory, provided one incorporates giant spins (clusters). Consequently, in what follows, we will refer only to reversible experiments and not field cooled ones [28]. No complete first principles theory has yet emerged for treating the irreversible contributions. However phenomenological approaches [12, 13] rely heavily on the notion that clusters of spins are important.

We have calculated the heat capacity $C_m$ and magnetization m in a finite magnetic field. This field enters the self-consistent equations for m, Q and M. We have numerically solved the three coupled equations for m, M and Q for a cluster of 10 spins with ferromagnetic exchange constant

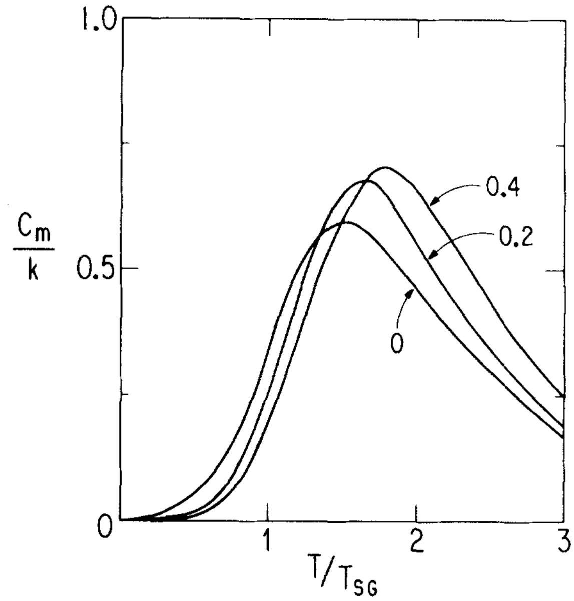



Fig. 3 Heat capacity $C_m$ versus temperature for fixed values of the magnetic field H in units of $g\mu_B/kT_{SG}$. The parameters are as defined in the text, $N = 10$, $\bar{J} = .01 J^o$ and $J' = .008 J^o$.

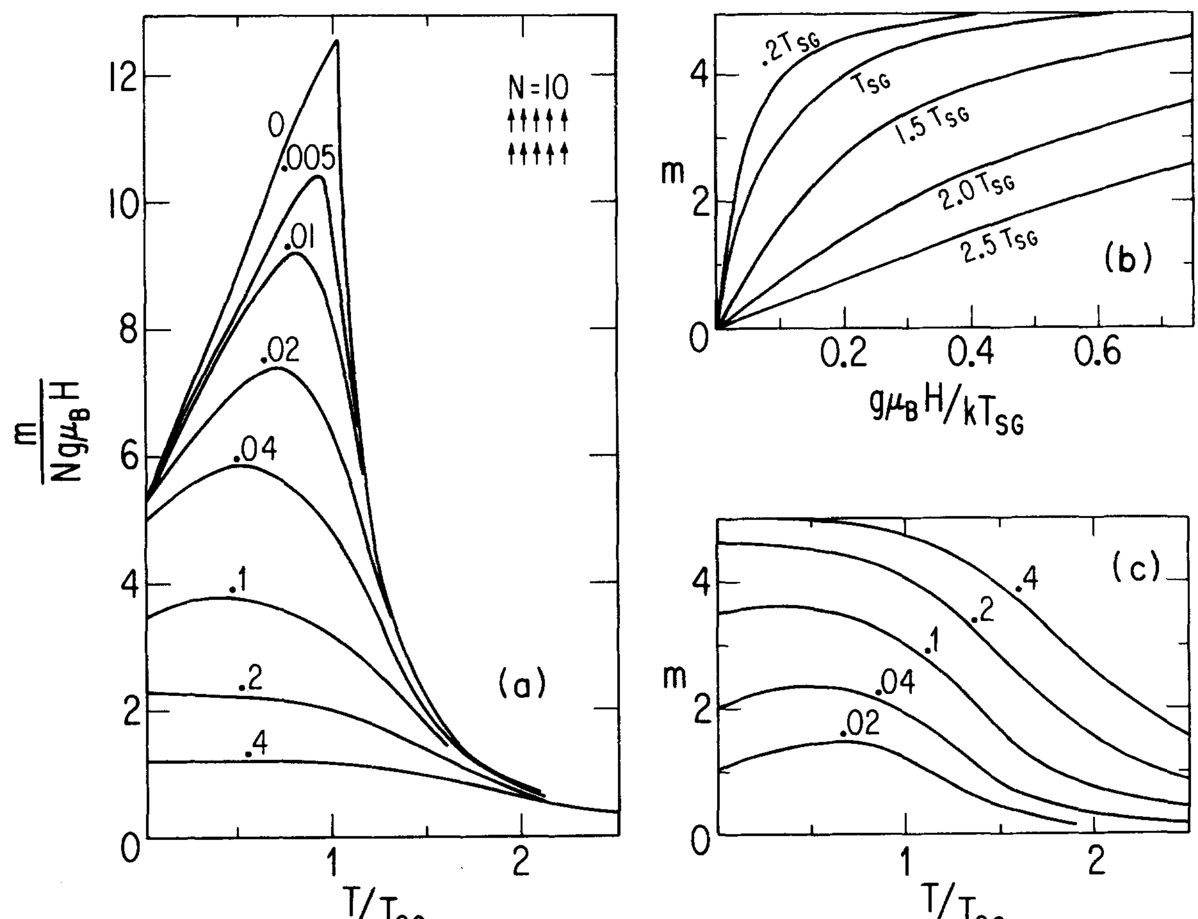



Fig. 4 Magnetization in a magnetic field H for 10 spins in a cluster. (a) $m/Ng\mu_B H$ versus T for several values of the field H in units of $g\mu_B H/kT_{SG}$. The curve denoted by 0 corresponds to $\chi = \frac{\partial m}{\partial H}\Big|_{H=0}$. (b) m versus H for fixed temperature T and (c) m versus T for several values of H in units of $g\mu_B H/kT_{SG}$.

$\bar{J}$ = .01 $J^o$ which is basically the width of the Gaussian (Eq. (2)) distribution for $P(J_{\mu\nu})$. We have also included a ferromagnetic inter-cluster exchange $J' = .008\ J^o$, to yield a relatively sharply peaked $\chi$ in zero field at $T_{SG}$= .236 $J^o$. In order to handle numerically large spin clusters we consider Ising spins. Our results for $\chi$ and m are only slightly affected by this simplification. However the specific heat Schottky anomaly is somewhat narrower than in the Heisenberg case.

The results for $C_m$ are shown for three values of the magnetic field (in units of $g\mu_B H/kT_{SG}$) in Fig. 3. As seen the effect of the field is to shift the maximum to higher temperatures; but with very little qualitative change in the shape of the curves. For this choice of parameters the inter-cluster contribution to $C_m$ (which yields a cusp at $T_{SG}$) is completely washed out by the Schottky anomaly. This need not always be the case, as shown in Refs. 1, 3.

By contrast the effects of the field on m and $\chi$ are more pronounced. In Fig. 4a, we plot m/H versus temperature for increasing values of field $g\mu_B H/k_B T_{SG}$ and for the same parameters as discussed above. The top curve denoted by 0 corresponds to $\chi = \partial m/\partial H|_{H=0}$. Note that relatively small fields both round and shift the maximum to lower temperatures. Fischer [18] showed that in the single spin EA model the cusp in $\chi$ becomes rounded when $g\mu_B H/k_B T_{SG} \sim 1$. Experimentally [9] the field dependence is about 20 to 50 times stronger than this. If the single spin EA model is replaced by a ferromagnetic cluster model containing 10 spins on average and $J' \neq 0$, the field sensitivity is increased by about a factor of 10-20. This leads to reasonable agreement with the reversible magnetization experiments, for AuFe alloys containing a few percent iron. In Fig. 4b, we plot m versus H for fixed T and in Fig. 4c m versus T for fixed H. From Fig. 4c, we find that for values of H below saturation, m has a maximum at a T approximately .5-.7 $T_{SG}$. This is also true for the single-spin EA model as m has a maximum in this temperature range for finite values of H. This is qualitatively similar to Beck's results on zero field cooled CuMn alloys. The experimental counterparts to Fig. 4a and c are given in Refs. [10, 16]. We find that our results compare qualitatively well with the zero-field cooled magnetization results.

G.S.G. wishes to acknowledge support from a Chiam Weizmann Postdoctorate Fellowship and K.L. wishes to acknowledge a grant from the Alfred P. Sloan Foundation. This work was supported by the NSF-Materials Research Laboratory.

REFERENCES

(a) Present address: Physics Department, Univ. of Virginia, Charlottesville, Virginia 22901.

1. C. M. Soukoulis and K. Levin, Phys. Rev. Lett. 39, 58 (1977); Phys. Rev. B 18, 1439 (1978).
2. C. M. Soukoulis, G. S. Grest and K. Levin, Phys. Rev. Lett. 41, 568 (1978).
3. C. M. Soukoulis, Phys. Rev. B, to be published.
4. L. E. Wenger and P. H. Keesom, Phys. Rev. B 11, 3497 (1975); ibid., B 13, 4053 (1976).
5. A. P. Murani et al., J. Phys. F 6, 425 (1976).
6. A. P. Murani, Phys. Rev. Lett. 37, 450 (1976); Proc. Conf. Neutron Scattering (Gatlinburg, Tennessee, 1976), Vol. 2, p. 657.
7. J. A. Mydosh, P. J. Ford, M. P. Kawatra, and T. E. Whall, Phys. Rev. B 10, 2845 (1974); P. J. Ford and J. A. Mydosh, ibid., B 14, 2057 (1976).
8. J. S. Schilling, P. J. Ford, V. Larsen, and J. A. Mydosh, Phys. Rev. B 14, 4368 (1976).
9. V. Cannella and J. A. Mydosh, Phys. Rev. B 6, 4220 (1976); V. Cannella, in Amorphous Magnetism, edited by H. O. Hooper and A. M. deGraaf (Plenum, New York, 1973), p. 195.
10. P. A. Beck, Met. Trans. 2, 2015 (1971); A. K. Mukopadhyay, R. D. Shull, and P. A. Beck, J. Less-Common Metals 43, 69 (1975).
11. A. P. Murani, J. Phys. F 4, 757 (1974).
12. J. L. Tholence and R. Tournier, J. Physique 35, C4-229 (1974); F. Holtzberg, J. L. Tholence, and R. Tournier, in Amorphous Magnetism II, edited by R. A. Levy and R. Hasegawa (Plenum, New York, 1976) p. 155.
13. C. N. Guy, J. Phys. F 7, 1505 (1977); ibid., F 8, 1309 (1978).
14. R. W. Knitter, J. S. Kouvel, and H. Claus, J. Magnetism Magnetic Mat. 5, 356 (1977).
15. J. A. Mydosh, A.I.P. 24, 131 (1975), in Amorphous Magnetism II, edited by R. A. Levy and R. Hasegawa (Plenum, New York, 1976), p. 73.
16. P. A. Beck, to be published.
17. S. F. Edwards and P. W. Anderson, J. Phys. F 5, 965 (1975).
18. K. H. Fischer, Phys. Rev. Lett. 34, 1438 (1975); Solid State Commun. 18, 1515 (1976).
19. D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. 35, 1792 (1975).
20. B. W. Southern, J. Phys. C 9, 4011 (1976).
21. K. Binder, Zeit. Phys. B 26, 339 (1977); in Festkorperprobleme XVIII, 55 (1977).
22. R. B. Coles, B. V. B. Sarkissian and R. H. Taylor, Phil. Mag. B 37, 489 (1978).
23. J. S. Kouvel, J. Appl. Phys. 31, 1425 (1960).
24. M. T. Beal-Monod, Phys. Rev. 178, 874 (1969); K. Matho and M. T. Beal-Monod, ibid. 135, 1899 (1972).
25. U. Larsen, Phys. Rev. B 14, 4356 (1976); J. Appl. Phys. 49, 1610 (1978).
26. K. Levin and D. L. Mills, Phys. Rev. B 9, 2354 (1974).
27. For definiteness, we took 10 spins in 2 x 5 clusters with $k_F a = 1.5$, where $k_F$ is the Fermi momentum and a is the lattice spacing. We used $T_K/T_{SG}$ = .01 and $\gamma = 2n(0)J^{sd} = -.08$, its value for AuFe.
28. W. Kinzel and K. H. Fischer, J. Phys. F 7, 2163 (1977).